

Application No. 09/937,599
Art Unit 1713
Tuesday, July 6, 2004
Reply to Office Action Dated March 3, 2004

AMENDMENTS TO THE SPECIFICATION

In the Abstract:

Please amend the Abstract as follows. A clean version of the amended Abstract is attached hereto at the end of this reply.

ABSTRACT

A kneading status evaluation method for a rubber composition containing at least a rubber and a filler ~~comprises~~ having the steps of a complex modulus measurement step (1) in which a complex modulus $E^*(a)$ at a given strain ϵ_a and a complex modulus $E^*(b)$ at a given strain ϵ_b differing from the strain ϵ_a of the rubber composition (I) are measured, a filler dispersion index calculation step (2) in which a filler dispersion index (N) of the rubber composition (I) is calculated with complex elastic moduli $E^*(a)$ and $E^*(b)$ obtained in the previous step (1), ~~according to the equation shown below,~~ and a comparison step (3) to compare a predetermined target filler dispersion index (R) with the filler dispersion index (N) calculated in the previous step (2), and/or a complex viscosity coefficient measurement step (5) to measure a complex viscosity coefficient η^* of the rubber composition (I) under at least two different temperatures, and a kneading status monitor index calculation step (6) to calculate a kneading status monitor index (M) of the rubber composition (I) ~~according to the equation shown below~~ on the basis of a temperature dependency of the complex viscosity coefficient

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η^* obtained at the previous step (5), and a comparison step (7) to compare a predetermined target kneading status monitor index (P) with the kneading status monitor index (M) calculated in the previous step (6). ~~(6).~~ The filler dispersion index (N) = $|E^*(a)|/|E^*(b)|$; $|\eta^*(T)| = A \exp (M/ RT)$, where η^* : complex viscosity coefficient, A: proportional constant, R: gas constant, and T: measuring temperature ($^{\circ}\text{K}$).

$$\text{Filler dispersion index (N)} = |E^*(a)|/|E^*(b)|$$

$$|\eta^*(T)| = A \exp (- M/ RT)$$

~~where η^* : complex viscosity coefficient, A: proportional constant, R: gas constant, and T: measuring temperature ($^{\circ}\text{K}$).~~

~~A manufacturing method for a rubber composition is characterized by carrying out the evaluation methods described above.~~

~~Implementation of the evaluation methods described above makes it possible to evaluate objectively a kneading status of a rubber composition containig at least a rubber and a filler. Further, implementation of the manufacturing methods described above can provide a rubber composition having good filler dispersion and a stable kneading status.~~

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In the Specification:

Please amend the paragraph at page 2, lines 10-17, as follows:

Hence, in the case of kneading an ethylene- α -olefin based ~~ting~~ copolymer rubber with a filler, a method of applying a large shear stress or prolonging a kneading time is conventionally adopted to achieve a fine filler dispersion, and then kneading conditions are adjusted by selecting more efficient kneading conditions of them with respect to product properties and processability in order to avoid lowering investment cost-performance and productivity.

Please amend the paragraph at page 2, lines 18-21, as follows:

However, there has been no definitive index for a kneading status and filler dispersion, and therefore, it is common at the present day that kneading conditions are decided according to ~~arbitral~~ arbitrary standards.

Please amend the paragraph beginning on page 2, line 16, as follows:

As a conventional method to evaluate filler dispersion, an electric resistance measurement method, a microscope method, and a light reflection method ~~basing~~ based on a degree of light reflected from a rubber compound surface are known so far. However, ~~a use of~~ using these methods as a monitoring index for detecting the defects described above

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is not sufficient because values given by these methods are subject to influence of a water content, a molecular weight distribution of polymer and so on, so that a definitive result can not be obtained even though the values are changeable somehow according to a variation of filler dispersion.

Please amend the paragraph beginning on page 4, line 23, as follows:

As a matter of fact, as an analytical method capable ~~to provide~~ of providing both a filler dispersion index and a kneading status monitor index at high accuracy, the wide range NMR method (Kiuchi Yasutaro and Ito Masayoshi: Japan Rubber Associate Magazine, 72, 1999) has been known. However, because of its high analysis cost and a slow response for an evaluation result, this method is not appropriate to use as an analytical method for quality control in a factory.

Please amend the paragraph at page 6, lines 3-7, as follows:

The present invention has a purpose of solving the problems described above which are inherent in conventional compounding technologies, and of providing evaluation methods to evaluate objectively a kneading status of a rubber composition containing at least a rubber and a filler.

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Please amend the paragraph beginning on page 10, line 5, as follows:

The second evaluation method for kneading status of the present invention is a kneading status evaluation method for a rubber composition (I) containing at least a rubber (A) and a filler (B), which comprises the steps of:

(5) a complex viscosity coefficient measurement step to measure a complex viscosity coefficient η^* of the rubber composition (I) under at least two different temperatures;

(6) a kneading status monitor index calculation step to calculate a kneading status monitor index (M) of the rubber composition (I) according to the following equation;

$$\cancel{|\eta^*(T)|} = \cancel{A \exp(-M/RT)} \quad \underline{|\eta^*(T)| = A \exp(M/RT)}$$

(where η^* : complex viscosity coefficient, A: proportional constant, R: gas constant, and T: measuring temperature ($^{\circ}\text{K}$)),

that shows a temperature dependency of the complex viscosity coefficient η^* obtained at the complex viscosity coefficient measurement step (5); and

(7) a comparison step to compare a predetermined target kneading status monitor index (P) with the kneading status monitor index (M) calculated in the kneading status monitor index calculation step (6).

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Please amend the paragraph beginning on page 11, line 13, as follows:

That is, this simple alternate is a kneading status evaluation method for a rubber composition (I) containing at least a rubber (A) and a filler (B), which comprises the steps of;

(5') a complex viscosity coefficient measurement step to measure a complex viscosity coefficient η' as a real part of complex viscosity coefficient η^* of the rubber composition (I) under at least two different temperatures;

(6') a kneading status monitor index calculation step to calculate a kneading status monitor index (M') of the rubber composition (I) according to the following equation;

$$\eta'(T) = A \exp(-M'/RT) \quad \eta'(T) = A \exp(M'/RT)$$

(where A: proportional constant, R: gas constant, and T: measuring temperature ($^{\circ}\text{K}$)),

that shows a temperature dependency of a real viscosity coefficient η' obtained as a real part of complex viscosity coefficient η^* at the complex viscosity coefficient measurement step (5'); and

(7') a comparison step to compare a predetermined target kneading status monitor index (P') with the kneading status monitor index (M') calculated in the kneading status monitor index calculation step (6').

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Please amend the paragraph beginning on page 16, line 14, and ending at page 17, lines 5, as follows:

(2) Reference kneading status monitor index (P): Temperature dependency of a complex viscosity coefficient of an uncured rubber composition obtained by kneading at least an ethylene- α -olefin based copolymer rubber with a reinforcing filler at a temperature of 100° or lower by using an 8-inch open roll mill without any vulcanizing agent, any cross-linking agent, any vulcanization accelerator, nor any cross-linking aid is shown in the following equation;

$$\eta^* = A \exp(-E_a / RT) \quad \eta^* = A \exp(E_a / RT)$$

(where η^* : complex viscosity coefficient, E_a : apparent activation energy, T : measuring temperature (°K), R : gas constant, and A : proportional constant), or

$$a_T = A \exp(-E_a / RT) \quad a_T = A \exp(-E_a / R(T - T_{ref.}))$$

(where a_T : shift factor, E_a : apparent activation energy, T : measuring temperature (°K), $T_{ref.}$: reference temperature, R : gas constant, and A : proportional constant).

WLF equation;

$$\eta_T = a_T \eta_{T0}$$

where a viscosity at the temperature T is designated η_T and a viscosity at a reference temperature T_0 is designated η_{T0} .

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Please amend the paragraph beginning on page 18, line 10, and ending on page 19, line 4, as follows:

(4) Kneading status monitor index (M): Temperature dependency of a complex viscosity coefficient of an uncured rubber composition (the same formulation as the uncured rubber composition described in (2)), which is obtained by kneading at least an ethylene- α -olefin based copolymer rubber with a reinforcing filler in a closed type mixer while a share stress is applied or both a share stress and heat are applied without any vulcanizing agent, any cross-linking agent, any vulcanization accelerator, nor any cross-linking aid, is shown in the following equation;

$$\eta^* = A \exp(-E_a / RT) \quad \eta^* = A \exp(E_a / RT)$$

(where η^* : complex viscosity coefficient, E_a : apparent activation energy, T : measuring temperature ($^{\circ}\text{K}$), R : gas constant, and A : proportional constant),

or

$$a_T = A \exp(-E_a / RT) \quad a_T = A \exp(-E_a / R(T - T_{\text{ref}}))$$

(where a_T : shift factor, E_a : apparent activation energy, T : measuring temperature ($^{\circ}\text{K}$), T_{ref} : reference temperature, R : gas constant, and A : proportional constant).

WLF equation;

$$\eta_T = a_T \eta_{T0}$$

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where a viscosity at the temperature T is designated η_T and a viscosity at a reference temperature T0 is designated η_{T0} .

Please amend the paragraph at page 30, lines 14-24, as follows:

Technological concept of this evaluation method is explained below. As is clear by the following equation, the M used in the equation shows temperature dependency of a complex viscosity coefficient η^* .

$$\cancel{|\eta^*(T)| = A \exp(-M/RT)} \quad |\eta^*(T)| = A \exp(M/RT)$$

In this equation, A is a proportional constant, R is the gas constant, and T is a measuring temperature ($^{\circ}\text{K}$). The temperature dependency of a complex viscosity coefficient η^* becomes ~~weaker~~ stronger according to increase of M value, in contrast, the temperature dependency of a complex viscosity coefficient η^* becomes ~~stronger~~ weaker according to decrease of M value.

Please amend the entire page 31 of the specification as follows:

An influence of pseudo-gel formation and deformation ~~in the rubber composition (I)~~ becomes ~~stronger~~ weaker according to an increase of temperature dependency of a complex viscosity coefficient η^* . The pseudo-gel formation and deformation scarcely occurs in the rubber composition (I) which is in a good kneading status, and hence the temperature dependency of a complex viscosity coefficient η^* becomes

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~~minimum~~ maximized while M value becomes ~~maximum~~ maximized. In other words, it can be said that the rubber composition (I) with a large M value is a rubber composition which is in a good kneading status, and the rubber composition (I) with a small M value is a rubber composition which is in a bad kneading status.

Consequently, the kneading status of the rubber composition (I) can be objectively evaluated by measuring a complex viscosity coefficient η^* at two or more different temperatures and by calculating M value from the Arrhenius plots.

For a reference, there are two calculation methods for M value. The M value can be obtained from the equation (1) or the equation (2) shown below. ~~The Ea~~ The Ea in these equations corresponds to the M value.

$$\cancel{|\eta^*| = A \exp(-E_a/RT) \dots (1)} \quad |\eta^*| = A \exp(E_a/RT) \dots (1)$$

In this equation (1), η^* is a complex viscosity coefficient, E_a is an apparent activation energy, T is a measuring temperature ($^{\circ}\text{K}$), R is the gas constant, and A is a proportional constant.

Please amend the paragraph on page 32, lines 1-4, as follows:

$$\cancel{a_T = A \exp(-E_a/RT) \dots (2)} \quad a_T = A \exp(-E_a / R(T-T_{\text{ref}})) \dots (2)$$

In this equation (2), a_T is a shift factor, E_a is an apparent activation energy, T is a measuring temperature ($^{\circ}\text{K}$), $T-T_{\text{ref}}$: reference temperature, R is the gas constant, A is a proportional constant.

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WLF equation;

$$\eta_T = a_T \eta_{T0}$$

where a viscosity at the temperature T is designated η_T and a viscosity at a reference temperature T0 is designated η_{T0} .

Please amend the paragraph at page 34, lines 12-20, as follows:

In the kneading status monitor index calculation step (6), on a bases of temperature dependency of complex viscosity coefficient η^* which is obtained at the complex viscosity coefficient measurement step (5), a kneading status monitor index (M) of the rubber composition (I) can be calculated according to the following equation;

$$\cancel{|\eta^*(T)| = A \exp(-M/RT)} \quad |\eta^*(T)| = A \exp(M/RT)$$

(where η^* : complex viscosity coefficient, A: proportional constant, R: gas constant and T: measuring temperature ($^{\circ}\text{K}$)).

Please amend the paragraph at page 42, lines 2-18, as follows:

(2) Reference kneading status monitor index (P): Temperature dependency of a complex viscosity coefficient of an unvulcanized rubber composition obtained by kneading at least an ethylene- α -olefin based copolymer rubber with a reinforcing filler at a temperature of 100°C or lower by using an 8-inch open roll mill without any vulcanizing agent, any cross-linking agent, any vulcanization accelerator, nor any cross-linking aid is shown in the following equation;

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$$\eta^* = A \exp(-E_a / RT) \quad \underline{\eta^* = A \exp(E_a / RT)}$$

(where η^* : complex viscosity coefficient, E_a : apparent activation energy, T : measuring temperature ($^{\circ}\text{K}$), R : gas constant, and A : proportional constant);

or

$$\underline{a_T = A \exp(-E_a / RT)} \quad \underline{a_T = A \exp(-E_a / R(T - T_{\text{ref}}))}$$

(where a_T : shift factor, E_a : apparent activation energy, T : measuring temperature ($^{\circ}\text{K}$), T_{ref} : reference temperature, R : gas constant, and A : proportional constant).

WLF equation;

$$\underline{\eta_T = a_T \eta_{T0}}$$

where a viscosity at the temperature T is designated η_T and a viscosity at a reference temperature T_0 is designated η_{T0} .

Please amend the paragraph beginning on page 44, line 13, and ending on page 45, line 6, as follows:

(4) Kneading status monitor index (M): Temperature dependency of a complex viscosity coefficient for an uncured rubber composition (the same formulation as the uncured rubber composition (2) described above), which is obtained by kneading at least an ethylene- α -olefin based copolymer rubber with a reinforcing filler in a closed type mixer while a share stress is applied or both a share stress and heat are applied without any vulcanizing agent, any cross-linking agent, any

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vulcanization accelerator, nor any cross-linking aid, is shown in the following equation;

$$\cancel{\eta^* = A \exp(-E_a / RT)} \quad \underline{\eta^* = A \exp(E_a / RT)}$$

(where η^* : complex viscosity coefficient, E_a : apparent activation energy, T : measuring temperature ($^{\circ}\text{K}$), R : gas constant, and A : proportional constant), or

$$\cancel{a_T = A \exp(-E_a / RT)} \quad \underline{a_T = A \exp(-E_a / R(T - T_{\text{ref}}))}$$

(where a_T : shift factor, E_a : apparent activation energy, T : measuring temperature ($^{\circ}\text{K}$), T_{ref} : reference temperature, R : gas constant, and A : proportional constant).

WLF equation;

$$\underline{\eta_T = a_T \eta_{T0}}$$

where a viscosity at the temperature T is designated η_T and a viscosity at a reference temperature T_0 is designated η_{T0} .